LANSCE
Los Alamos Neutron Science Center

Research Proposal for the use of Neutron Science Facilities

☐ **Fast Access** ☐ **Joint CINT Proposal**

Proposal Number: 20111554
Submission Number: S1574
Date Received: 03/14/11

Program Advisory Subcommittee: Nuclear Technology
Focus Area:

Flight Path/Instrument: Target 2 / Blue Room
Estimated Beam Time (days): 15
Days Recommended: 0
Dates Desired: Split:
Impossible Dates:

TITLE
Lead Slowing-Down Spectrometer Experiments

☒ Continuation of Proposal #: 20101554
☐ Ph.D Thesis for:

Principal Investigator: Haight, Robert C
Institution: Los Alamos National Laboratory
Citizenship: United States of America
Phone: 505-667-2829 **FAX:** 505-665-3705
Email: haight_robert_c@lanl.gov
Local Contact: Bitteker, Leo J

Co-Proposers	Institution	Citizenship	E-mail Address
Belier, Gilbert	Commissariat a l'Energie Atomic	France	gilbert.belier@cea.fr
Bredeweg, Todd A	Los Alamos National Laboratory	United States of Am	toddb@lanl.gov
Danon, Yaron	Rensselaer Polytechnic Institute	Israel	danony@rpi.edu
Devlin, Matthew	Los Alamos National Laboratory	United States of Am	devlin@lanl.gov
Gavron, Victor	Los Alamos National Laboratory	United States of Am	gavron@lanl.gov
Jandel, Marian	Los Alamos National Laboratory	Slovak Republic	mjandel@lanl.gov
O'Donnell, John M	Los Alamos National Laboratory	United Kingdom	odonnell@lanl.gov
Thompson, Jason T	Rensselaer Polytechnic Institute	United States of Am	.

RESEARCH AREA

- ☐ Biological and Life Science
- ☐ Chemistry
- ☐ National Security
- ☐ Earth Sciences
- ☐ Engineering
- ☐ Environmental Sciences
- ☒ Nuc. Physics/chemistry
- ☐ Astrophysics
- ☐ Few Body Physics
- ☐ Fund. Physics
- ☐ Elec. Device Testing
- ☐ Dosimetry/Med/Bio
- ☐ Earth/Space Sciences
- ☐ Materials Properties/Test
- ☐ Other:
- ☐ Mat'l Science (incl Cond Matter)
- ☐ Medical Applications
- ☐ Nuclear Physics
- ☐ Polymers
- ☐ Physics (Excl Condensed Matter)
- ☒ Instrument Development
- ☒ Neutron Physics
- ☒ Fission
- ☒ Reactions
- ☐ Spectroscopy
- ☒ Nuc. Accel. Reactor Eng.
- ☒ Def. Science/Weapons Physics
- ☐ Radiography
- ☐ Threat Reduction/Homeland Sec.
- ☐ Other:

FUNDING AGENCY

- ☐ DOE/BES
- ☐ DOE/OBER
- ☒ DOE/NNSA
- ☒ DOE/NE
- ☐ DOE/SC
- ☐ DOE/Other
- ☐ DOD
- ☐ NSF
- ☐ Industry
- ☐ NASA
- ☐ NIH
- ☐ Foreign:
- ☐ Other US Gov't:
- ☐ Other:

PUBLICATIONS

Publications:

DANCE: A. M. Alpizar-Vicente, T. A. Bredeweg, E.-I. Esch, U. Greife, R. C. Haight, R. Hatarik, J. M. O'Donnell, R. Reifarth, R. S. Rundberg, J. L. Ullmann, D. J. Vieira, and J. M. Wouters, "Neutron capture cross section of 62Ni at s-process energies," Phys. Rev. C 77, 015806 (2008).

DANCE: S. A. Sheets, U. Agvaanluvsan, J. A. Becker, F. Bečvář, T. A. Bredeweg, R. C. Haight, M. Krtička, M. Jandel, G. E. Mitchell, J. M. O'Donnell, W. E. Parker, R. Reifarth, R. S. Rundberg, E. I. Sharapov, I. Tomandl, J. L. Ullmann, D. J. Vieira, J. M. Wouters, J. B. Wilhelmy, and C. Y. Wu, "Spin and parity assignments for 94,95Mo neutron resonances," Phys. Rev. C 76, 064317 (2007)

FIGARO: T. Ethvignot, M. Devlin, H. Duarte, T. Granier, R. C. Haight, B. Morillon, R. O. Nelson, J. M. O'Donnell, and D. Rochman, "Reply to Comment on 'Neutron Multiplicity in the Fission of 238U and 235U with Neutrons up to 200 MeV' " Phys. Rev. Lett. 101, 039202 (2008).

DANCE: M. Jandel, T. A Bredeweg, E. M. Bond, M. B. Chadwick, R. R. Clement, A. Couture, J. M. O'Donnell, R. C. Haight, T. Kawano, R. Reifarth, R. S. Rundberg, J. L. Ullmann, D. J. Vieira, J. B. Wilhelmy, J. M. Wouters, U. Agvaanluvsan, W. E. Parker, C. Y. Wu and J. A. Becker, "Neutron Capture Cross Section of 241Am," LA-UR-08-03246 (2008). Phys. Rev. C 78, 034609 (2008)

FIGARO: R. C. Haight, S. Nelson and J. M. O'Donnell, "Los Alamos Analysis of an Experiment to Measure

Abstract: S1574_Haight_LSDS_.doc

By electronic submission, the Principal Investigator certifies that this information is correct to the best of their knowledge.

Safety and Feasibility Review *(to be completed by LANSCE Instrument Scientist/Responsible)*

☐ **No further safety review required** ☐ **To be reviewed by Experiment Safety Committee**

☐ **Approved by Experiment Safety Committee, Date:**

Recommended # of days:	**Change PAC Subcommittee and/or Focus Area to:**	**Change Instrument to:**

Comments for PAC to consider:

Instrument scientist signature: **Date:**

Lead Slowing-Down Spectrometer Experiments

R. C. Haight, H. Y. Lee, J. M. O'Donnell, M. Devlin, A. Laptev (LANSCE-NS)

D. J. Vieira, T. A. Bredeweg, M. Jandel (C-NR)

V. Gavron, (LANSCE-DO)

Y. Danon, J. Thompson (RPI)

G. Belier (CEA)

Proposal for Beam Time in 2011

This proposal is an Omnibus Proposal for LSDS Experiments. These experiments include (in approximate order of priority)

1. Measurement of the neutron-induced fission cross section of 237U
2. Test of detectors for assaying nuclear fuel: detectors "blind" to neutrons below 10 keV are required.
3. Thesis research for RPI graduate student
4. (n,p) and (n,alpha) development

Two of these measurements (#1 and #2) have very high priority in order to meet programmatic milestones. Because the preparation of the 237U sample (6.7 day half-life) requires an irradiation at the High Flux Isotope Reactor at ORNL, the scheduling of this measurement must take into account the HFIR schedule and the time necessary to process the material at ORNL, install it in a shielded fission chamber, and then to ship it to us at LANSCE.

The reasons for the measurements are as follows:

1. **<u>237U(n,f)</u>** – This isotope has a half-life of 6.75 days and can be produced by 238U(n,2n) reactions in reactors and nuclear explosions. The neutron-induced fission cross section has been measured with neutrons at thermal, at 200 keV and with neutrons from a nuclear explosion. The experimental data are shown in Fig. 1.

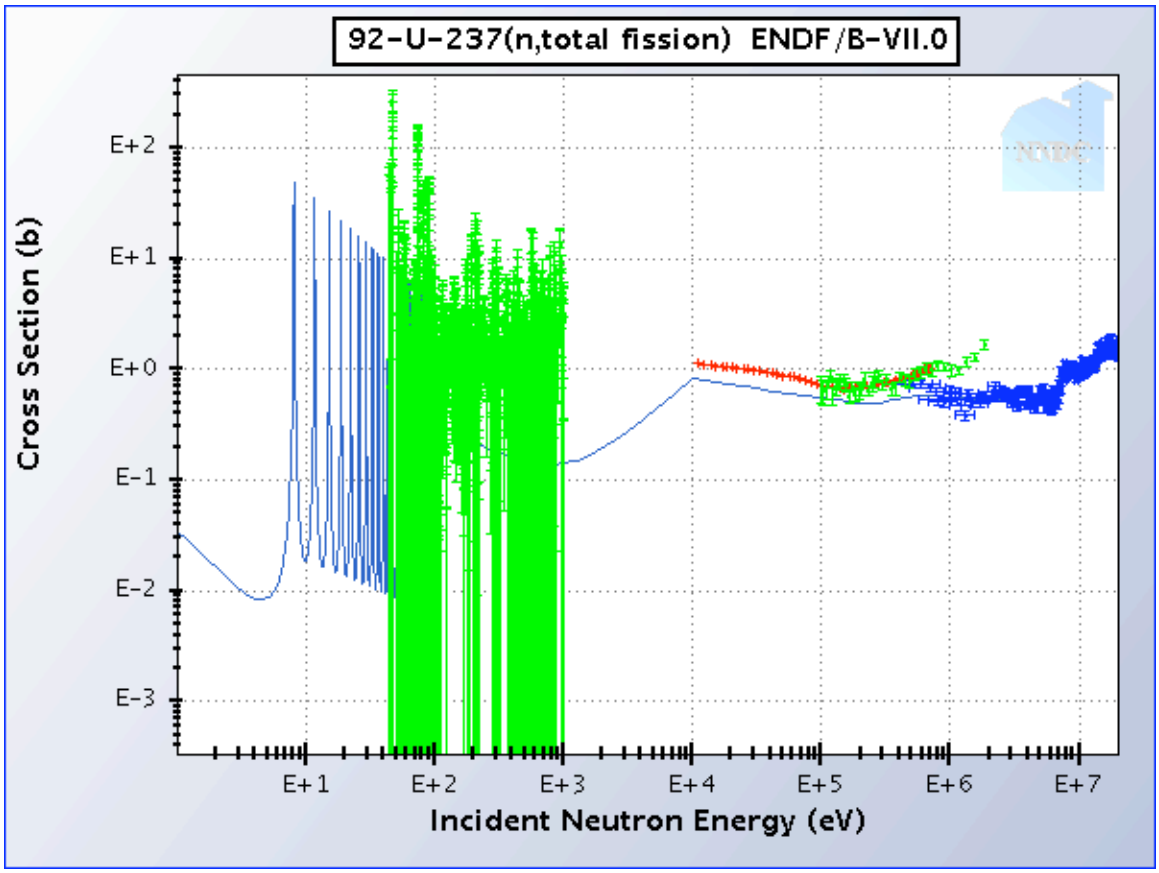


Fig. 1 – 237U(n,f) cross section: Blue is ENDF/B-VII.0 evaluation; green are points from McNally from a nuclear explosion neutron source (1974); red points are an evaluation from Caner (1976); the black point at thermal is from Cowen (1955); and the blue points are from surrogate reactions by Burke (2006). Note that there are no experimental data above thermal to 44 eV and from1 keV to 100 keV.

The method of making the sample is to take a highly enriched sample of 236U and subject it to a high thermal neutron flux to produce 237U by the 236U(n,gamma) reaction. In addition, the thermal neutron flux will burn out the 235U impurity. The proposal by Jerry Wilhelmy is attached to this proposal (Appendix A).

In 2010, we had our first run with a sample of 237U prepared at HFIR. A significant signal from 237U(n,f) was observed. We now need to quantify the background better from 236U(n,f). The sample and detector have not been removed from the LSDS and so we can measure the background by a run in June, 2011.

Following this run, we propose measuring a second sample of 237U, with higher purity and more mass. We would choose an upper-level channel in the LSDS to minimize scattering from the aluminum supporting base plate. This run would be followed by a third run to measure the background in this sample.

First run – 1 day linac followed by 4 days PSR:

The exact dates of this run are not crucial as it is with the sample and detector used last year. The purpose is to quantify background fissions from isotopes other than 237U in that sample. We suggest June 22-26, 2011 with previously run sample and detector.

Second run – 1 day linac followed by 4 days PSR:

The dates are crucial: they depend on the HFIR irradiation schedule (Appendix B) and the possible LSDS running schedule. The present schedules of each are in Fig. 2. The LSDS run would need to begin 20 days after the start of the HFIR irradiation, and that irradiation would need to be about 2 half-lives (2 x 6.7 = 13 days) long. The present HFIR preferences are to perform the irradiation in August or October. Here is a brief analysis of possible dates:

a. August does not seem to be a possibility as the sole use time is only 18 days after the beginning of the HFIR irradiation time that month.
b. October is out unless the LSDS run is in early November (e.g. Nov. 1-5). But then moving the LSDS immediately out of the beam line so that Target 4 could run would risk high radiation doses to personnel.
c. December 16-21 looks good. HFIR irradiation would start about November 26.

Some consideration should be given to modifying the LANSCE running schedule to accommodate this important, second run.

Third run – 1 day linac followed by 4 days PSR:

The dates are not crucial but they need to be at least one week following run #2. They could be several months later. We would quantify the backgrounds in run #2.

LANSCE schedule + HFIR (→)

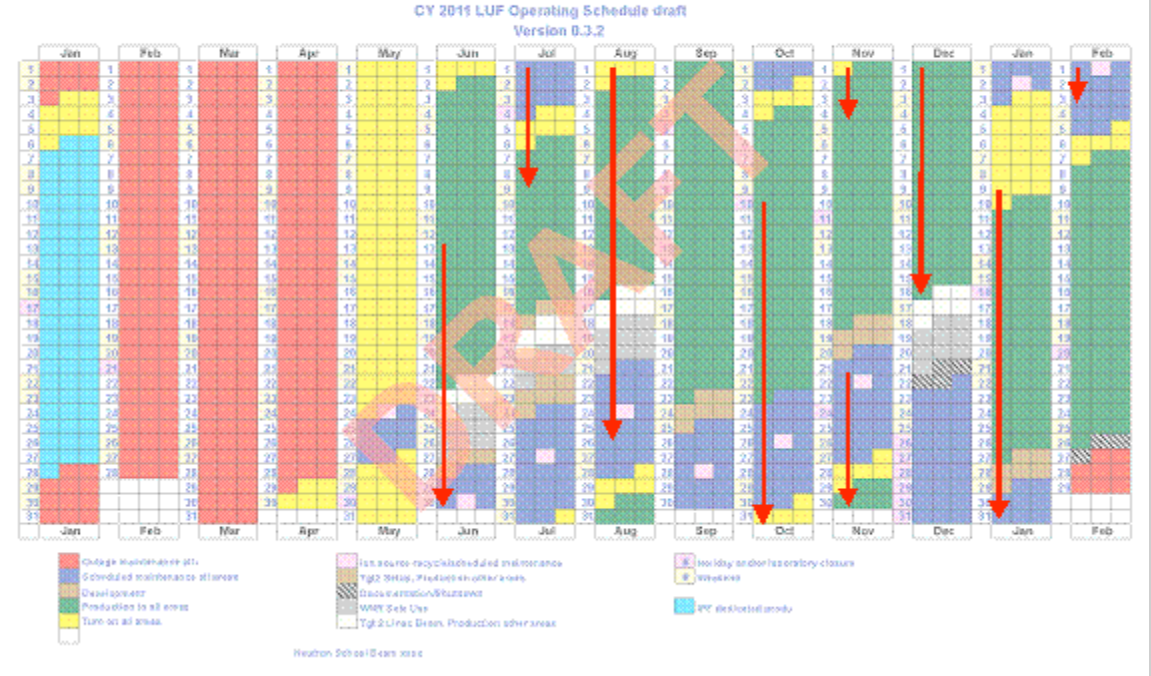


Fig. 2 - LANSCE and HFIR schedules through February, 2012. The irradiation to produce the 237U sample needs to be about 13 days long. The LSDS run needs to start 20 days after the beginning of the HFIR irradiation.

2. **<u>Test of detectors for assaying nuclear fuel: detectors "blind" to neutrons below 10 keV</u>**: The idea here is to assay spent fuel for 235U and 239Pu content by putting samples in the LSDS and looking for fast neutrons produced by (n,f) reactions in the resonance range. We had a successful experiment with 235U in 2010. Now we need to measure 239Pu.

The detectors proposed for the fast neutrons are fission chambers containing pure 232Th, which has a very low fission cross section for neutrons below 100 keV. The relative amounts of 235U and 239Pu in a complex sample would be determined by the resonant fission cross sections. We place a sample of 235U or 239Pu in the LSDS and look for fission resonances with the fast neutron detector.

This research is led by V. Gavron (LANSCE-DO).

3.-4. (n,p) and (n,alpha reactions:

These reactions have three motivations: (a) thesis research for Jason Thompson of RPI; (b) developing techniques to measure 73As(n,p) and (n,alpha) as part of an LDRD/DR project and 50V(n,p) and (n,alpha), all of which have application to defense programs; and (c) developing techniques to measure these reactions on 22Na, 26Al and 40K, which are important for astrophysical nucleosynthesis. Appendix C gives details of these measurements.

Measurements of (n, alpha) cross section are challenging because of the lower Q value of the reaction that result in a lower energy deposition in the detector. In addition, the intense neutron flux and electromagnetic background require development of detectors that have fast recovery and are insensitive to the background. Such effort is current underway at Rensselaer Polytechnic Institute (RPI) and is supported by a Stewardship Science Academic Alliance grant. As part of this work several detector concepts were developed including a compensated ion chamber, a compensated solar cell detector, a cylindrical ion chamber, a Gas Electron Multiplier (GEM) based detector and a Passivated Implanted Planar Silicon (PIPS) detector. We are currently focusing on the last two concepts which yielded good results in preliminary experiments with the RPI and LANL LSDS. Previous accomplishments include (n,alpha) measurements on ~9 mg of Sm-147 and Sm-149 with sub mb cross section (in part of the energy range). In this run we plan to measure V-50(n,p)Ti-50 with a proton energy of about 4.5 MeV and a theoretical estimate of the average cross section of 0.7mb. Enriched V-50 is on order and the sample will be made at RPI. This measurement is an important objective of our SSAA grant to demonstrate the feasibility of (n,alpha) and (n,p) measurements using the LSDS in preparation for radioactive sample measurement on materials of interest to the stockpile stewardship program. It is also an important milestone in Jason Thompson PhD thesis research.

SUMMARY

The beam request is therefore for 3 runs, each of 5 days. The first day of each run could be linac beam. The remaining 4 days each would be PSR-to-Blue Room runs. At least one setup day prior to the run would be required. After the run, the Blue Room should be allowed to cool down several days before the LSDS can be moved and other experiments, including those at Target 4, resumed.

APPENDIX A

^{237}U(n,f)

Jerry Wilhelmy

October 17, 2005

Goal: Measure ^{237}U(n,f) cross section in the 1 eV – 100 keV range
Using the lead slowing down spectrometer

^{237}U can be produced copiously in a nuclear device through (n,2n) reactions on ^{238}U. ^{237}U has a substantial fission cross section for neutrons at bomb thermal energies. Thus the fission yield from ^{237}U can be an important perturbation in the device environment. Previous experimental measurements of the ^{237}U(n,f) cross section have used neutrons generated with a Cockroft-Walton accelerator at around 200 keV(1), critical assembly exposures(2) and underground nuclear explosions(3). In addition, recently Younes and Britt(4) have reanalyzed "surrogate" data and presented an evaluated cross section for the ^{237}U(n,f) reaction.

^{237}U Production

^{237}U can be produced by bombarding ^{236}U in a high flux reactor. The experiments done in the 1970's irradiated 42mg of highly enriched ^{236}U in the HFIR at ORNL. The flux was 2.2x10^{15} n/cm^2-sec and the irradiations were performed over a 22 day reactor cycle. Under these conditions they reported that 1.9% of the ^{236}U was converted to ^{237}U.

Currently HFIR operates at 85 MW (in the 1970's it was 100 MW). The amount of ^{237}U (=N_7) produced at HFIR at EOB can be determined using the following equations:

$$\frac{dN_7}{dt} = \text{Rate of production} - \text{Rate of decay}$$

which gives on integration:

$$N_7 = \frac{\sigma_6 \phi N_6^0}{(\lambda_7 + \sigma_7\phi - \sigma_6\phi)}\left[e^{-\sigma_6\phi t} - e^{-(\lambda_7+\sigma_7\phi)t}\right] \qquad (Eq.1)$$

Where

N_7 = Atoms ^{237}U
σ_6 = ^{236}U(n,γ) cross section (cm^2)
σ_7 = ^{237}U(n,γ) cross section (cm^2)
ϕ = Reactor neutron flux (n/cm^{2}sec)
N_6^0 = Atoms ^{236}U at start of irradiation
λ_7 = ^{237}U decay constant (sec^{-1})

t = Reactor irradiation time (sec)

Dependant on the capture cross section and the details of the neutron fluence it may be desirable to explicitly consider both the thermal and resonant integral fluences and cross sections. Under these conditions (Eq 1) becomes:

$$N_7 = \frac{(\sigma_{T6}\phi_T + \sigma_{R6}\phi_R)N_6^0}{(\lambda_7 + (\sigma_{T7}\phi_T + \sigma_{R7}\phi_R) - (\sigma_{T6}\phi_T + \sigma_{R6}\phi_R))}\left[e^{-(\sigma_{T6}\phi_T + \sigma_{R6}\phi_R)t} - e^{-(\lambda_7 + \sigma_{T7}\phi_T + \sigma_{R7}\phi_R)t}\right] \quad (Eq.2)$$

Where

N_7 = Atoms ^{237}U
σ_{T6} = ^{236}U(n,γ) thermal cross section (cm^2)
σ_{R6} = ^{236}U(n,γ) resonance integral cross section (cm^2)
σ_{T7} = ^{237}U(n,γ) thermal cross section (cm^2)
σ_{R7} = ^{237}U(n,γ) resonant integral cross section (cm^2)
ϕ_T = Reactor thermal neutron flux (n/cm^{2}sec)
ϕ_R = Reactor resonant integral neutron flux (n/cm^{2}sec)
N_6^0 = Atoms ^{236}U at start of irradiation
λ_7 = ^{237}U decay constant (sec^{-1})
t = Reactor irradiation time (sec)

The relevant HFIR operation and nuclear reaction parameters are:

Parameter	Description	Value
σ_{T6}	^{236}U(n,γ) thermal cross section	5.1 barns
σ_{R6}	^{236}U(n,γ) resonance integral cross section	360 barns
σ_{T7}	^{237}U(n,γ) thermal cross section	400 barns
σ_{R7}	^{237}U(n,γ) resonant integral cross section	1200 barns
ϕ_T	Reactor thermal neutron flux	1.87x10^{15} n/cm^{2}sec
ϕ_R	Reactor resonant integral neutron flux	8.5x10^{13} n/cm^{2}sec
N_6^0	Atoms ^{236}U at start of irradiation	2.55x10^{18} (= 1 mg)
λ_7	^{237}U decay constant	1.19x10^{-6} sec^{-1} (=$t_{1/2}$ 6.75d)
t	Reactor irradiation time	1.9x10^6 sec (= 22d)

For reactor feed material, highly enriched ^{236}U is available with the following isotopic distribution:

Isotope	Abundance (%)
^{233}U	0.082
^{235}U	0.205

^{236}U	99.679
^{238}U	0.034

Using 1 mg of ^{236}U feed material we will have at the end of the HFIR irradiation cycle:

Isotope	Atoms (EOB)	Micrograms (EOB)
^{235}U	$4.33x10^{14}$	0.169
^{236}U	$2.36x10^{18}$	925.
^{237}U	$4.64x10^{16}$	18.3

Note: a 22day irradiation in HFIR will appreciably reduce (burn up) the initial ^{235}U feed. The 1 mg ^{236}U sample goes from an ingoing concentration of 2.05 μg of ^{235}U down to 0.17 μg at EOB (a reduction of ~ a factor of 12).

LSDS fission measurements

The fission rate obtained in the LSDS for the relevant U isotopes can be estimated using:

- ENDF (n,f) cross sections for $^{235,236}U$.
- The McNally(3) estimates of the ^{237}U(n,f) cross section
- LSDS neutron fluence @ 30 cm irradiation port for 1 μA proton beam.

The results are:

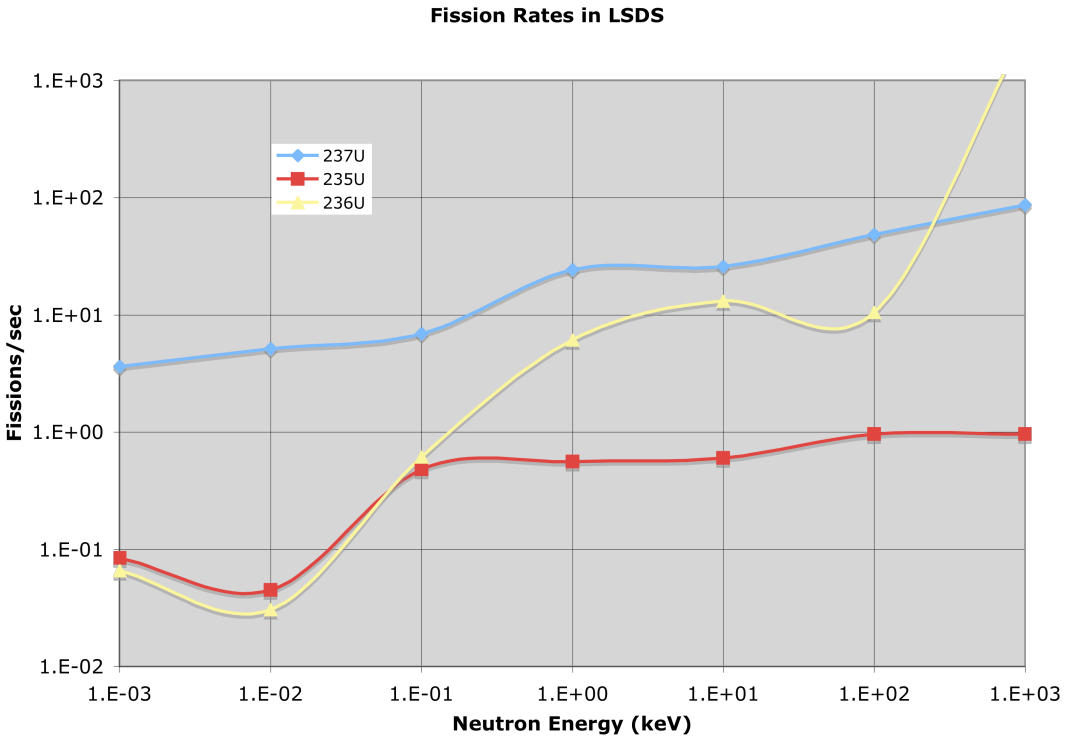


Fig 1

So over all energy ranges below ~ 200 keV the major fission rate component in the LSDS will be desired ^{237}U(n,f). However, it should be pointed out that the data in Fig 1 are for the isotopic distribution at the EOB of the reactor cycle. Given that the sample has to be returned to LANL and that clean up chemistry has to be preformed to remove other actinides and fission products it realistically will probably take on the order of a half life (~ 1 week) to get the sample into the LSDS for data acquisition. Even so, accurate background corrections will be possible by, after completion of the initial set of measurements, allowing the sample to decay for a few ^{237}U half lives and remeasuring the fission rates from the background component isotopes.

^{236}U(n,f)

The sub MeV ^{236}U(n,f) cross section is poorly known. What the nuclear data libraries carry seems to be just a downward scaling of the capture cross section. This is probably due to the fact that it is difficult to measure this small, thresholding cross section in the presence of even small ^{235}U contaminants in the target material. Even using the highly enriched ^{236}U feed material we have available to us, the low neutron induced fission rate will be dominated by the 0.2% ^{235}U content. I think this gives us an opportunity to make the first high quality measurements of this cross section. If, following HFIR irradiation and after the ^{237}U decays away we will have an even more highly depleted ^{235}U sample (~ 0.017% ^{235}U content). The ratio of neutron induced fission events in ^{236}U/^{235}U are shown in Fig. 2 for both before and after sample irradiation in HFIR.

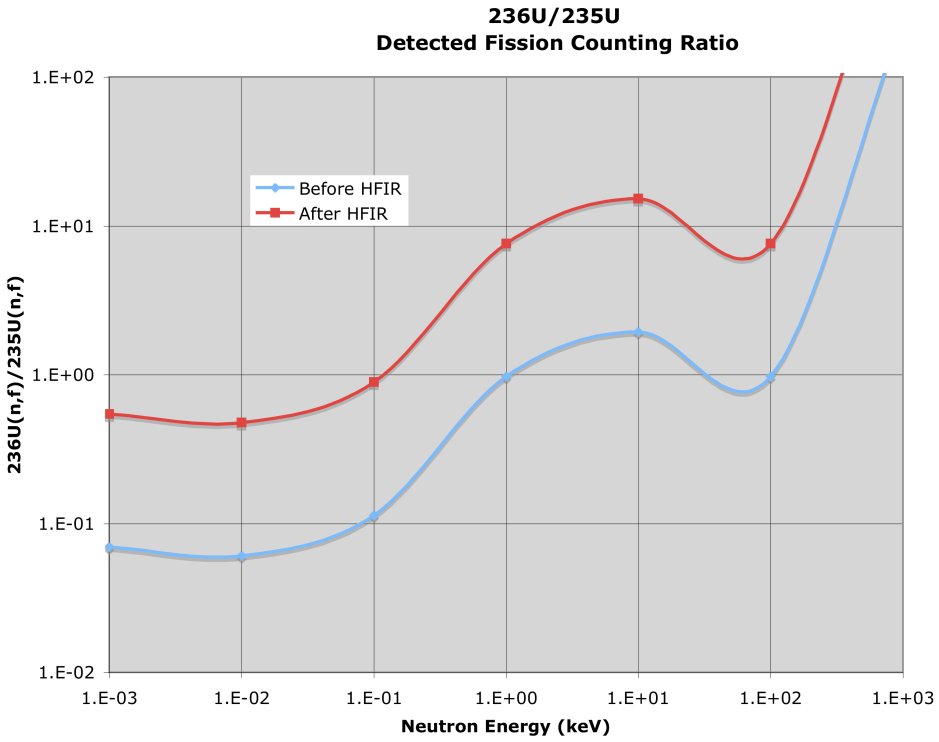


Fig. 2

As seen in the figure, the HFIR burn up of the ^{235}U will allow much better S/N for determining the ^{236}U(n,f) cross section in the sub MeV neutron energy range.

^{236}U(n,f) in DANCE?

Using the new fission detector trigger capabilities in DANCE it would be desirable to measure ^{236}U(n,f) in this facility. The problem is that the sub threshold fission cross section for ^{236}U(n,f) is (poorly) estimated to be in the 10^{-27} – 10^{-26} cm^2 range for eV through keV neutrons. These are very small cross sections for DANCE to measure especially since we also have to actively detect the fission event to have a trigger.

The detected fission rate in DANCE can be estimated as:

$$F = N_6 * \sigma_{(n,f)} * \phi * \Omega * t$$

where

F = observed ^{236}U(n,f) fission events
N_6 = Number of ^{236}U target atoms
$\sigma_{(n,f)}(E)$ = ^{236}U(n,f) cross section at neutron energy E (cm^2)
ϕ = neutron flux on target (n/(cm^2-sec))
Ω = efficiency for detecting fission events
t = irradiation time (sec)

N_6 is limited to the range for fission events to escape from the target. The maximum rate we can obtain (using a single target) will be to use a sample that is the thickness of the range of the fission products. The range of a light fission product (e.g. Kr) is ~ 15 mg/cm^2 in U (ref 5) and about 11 mg/cm^2 for a heavy fission product (e.g. Ba).

$\sigma_{(n,f)}(E)$ is taken for illustrative purposes to be 1 mb which is consistent with the values in the ENDF tabulations (ref 6).

ϕ is taken from Ullmann to be

ϕ = (7.0 x 10^3 N/cm^2/ev/sec)E(eV)

for a primary proton beam of 55 μA. This gives when integrated for a 10% ΔE/E measurement window a flux on target of ~ 700 N/(cm^2-sec).

Ω is the efficiency for fission fragments to emerge from the target. If we use a thick target of ~15 mg/cm^2 ^{236}U then we will have ~ 20% of the light fragments and ~ 13% of the heavy fragments exiting the target. This gives an overall efficiency of ~ 33% for having a fragment exit the target. If we use the most optimistic estimate that ALL of these will be detected in the DANCE fission detector then we will have the maximal detection efficiency.

"t" is the time of irradiation and for illustrative purposes is taken to be 1 day (i.e. 86,400 sec).

Combining these factors together we obtain

F ~ 0.76 detected fission events/(day-energy interval-mb)

This is clearly below the feasibility range. The above estimates also ignore the fissions that would be due to the small ^{235}U contaminant. As shown above this is an important perturbation unless substantial burn up of the ^{235}U is accomplished before DANCE irradiation. So it appears that it will not be possible to simply use DANCE to measure the $^{236}U(n,f)$ cross section over the eV – 100 keV range. However, it may be possible to use DANCE to look in the resonance region where fission enhanced resonances could occur where the cross section would be substantially above the 1 mb used in the above example.

As shown above, the best bet for measuring the $^{236}U(n,f)$ appears to be to use the LSDS on a sample that has been pre irradiated in a high flux reactor to further deplete the ^{235}U content.

Summary

1) $^{237}U(n,f)$ can be measured using the LSDS. Though the measurement appears to be pretty straight forward from a physics perspective there will be a tremendous amount of logistics necessary to bring this off in a correct time sequence.
    - High flux reactor irradiation
    - Chemical processing to purify the residual U from other capture and fission products.
    - Meshing with the LANSCE schedule
    - All ES&H approvals and work authorization packages
2) $^{236}U(n,f)$. In the sub MeV region this is a poorly known cross section. Following a high flux reactor irradiation of the highly enriched ^{236}U sample the small amount of ^{235}U will be further substantially depleted. This should provide a very good target to do these measurements.
    - A measurement in the LSDS can be done after sufficient time to let the ^{237}U decay away.
    - Due to the lower neutron fluence available at DANCE it appears difficult to attempt $^{236}U(n,f)$ measurements over the entire range of interest. However, it may be possible to gain new information in the neutron resonance region where there may be resonances with substantial fission cross sections. An exploratory study with DANCE and the fission detector capability seems warranted.

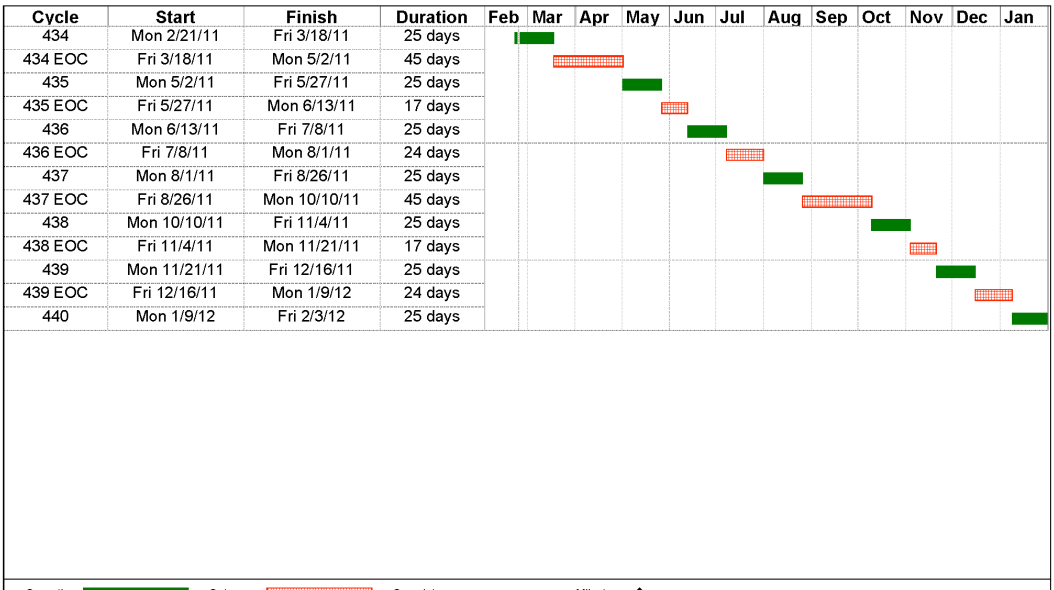
References:

1. G.A. Cowan, G.A. Jarvis, G.W. Knobeloch, and B. Warren, LASL Report LA-1669, 1955
2. D.W. Barr (private communication)

3. H.H. McNally, J.W. Barnes, B.J. Dropesky, P.A. Seeger, and K. Wolfsberg, Phys. Rev. C**9**, 717 (1974).
4. W. Younes and H.C. Britt, Phys. Rev. C**68**, 034610 (2003).
5. L.C. Northcliffe and R.F. Schilling, Nucl Data A**re**, 233 (1970).
6. http://www.nndc.bnl.gov/exfor/endf00.htm

APPENDIX B

Cycle	Start	Finish	Duration
434	Mon 2/21/11	Fri 3/18/11	25 days
434 EOC	Fri 3/18/11	Mon 5/2/11	45 days
435	Mon 5/2/11	Fri 5/27/11	25 days
435 EOC	Fri 5/27/11	Mon 6/13/11	17 days
436	Mon 6/13/11	Fri 7/8/11	25 days
436 EOC	Fri 7/8/11	Mon 8/1/11	24 days
437	Mon 8/1/11	Fri 8/26/11	25 days
437 EOC	Fri 8/26/11	Mon 10/10/11	45 days
438	Mon 10/10/11	Fri 11/4/11	25 days
438 EOC	Fri 11/4/11	Mon 11/21/11	17 days
439	Mon 11/21/11	Fri 12/16/11	25 days
439 EOC	Fri 12/16/11	Mon 1/9/12	24 days
440	Mon 1/9/12	Fri 2/3/12	25 days



Operation Outage Complete Milestone

HFIR

12 Month Planning Schedule

Operating schedule length is normally more than 22 days but not exceeding 27 days. For planning purposes, a 25 day schedule length is shown. This schedule is for planning purposes only and is subject to change.

APPENDIX C

Measurements of (n,p) and (n.alpha) cross section for small samples using the LANL Lead Slowing Down Spectrometer

Y. Danon and J. Thompson
Rensselaer Polytechnic Institute, Troy, NY 12180

1. Introduction

The Los Alamos National Laboratory (LANL) Lead Slowing Down Spectrometer (LSDS) is a unique instrument providing high neutron flux that enables measurements of neutron induced cross section for very small samples (nanograms) or for samples with the very low cross sections (microbarns).

Utilization of this instrument requires development of detectors suitable for operation in this high flux environment. Previous work with the LANL LSDS aimed at measurement of the fission cross section of ^{235m}U resulted in a successful proof of principle measurement of ^{239}Pu fission cross section with sample size of less than 10 nanograms[i].

Additional opportunity exists in utilizing the LSDS for measurements of (n,α) and (n,p) cross sections on small samples, radioactive samples or samples with very low cross section. Such measurements have applications in astrophysics, reactor applications and stockpile stewardship.

Measurements of (n,α) cross section are more challenging because of the lower *Q* value of the reaction that result in a lower energy deposition in the detector. In addition, the intense neutron flux and electromagnetic background require development of detectors that have fast recovery and are insensitive to the background. Such effort is current underway at Rensselaer Polytechnic Institute (RPI) and is supported by a Stewardship Science Academic Alliance grant. As part of this work several detector concepts were developed including a compensated ion chamber, a compensated solar cell detector, a cylindrical ion chamber, a Gas Electron Multiplier (GEM) based detector and a Passivated Implanted Planar Silicon (PIPS) detector. We are currently focusing on the last two concepts which yielded good results in preliminary experiments with the RPI LSDS.

Tests with the RPI LSDS provide initial data about the detectors and can also be used to perform actual measurements. The LANL LSDS has a neutron flux which is about 2 orders of magnitude higher than the RPI LSDS and thus enable measurement on smaller samples. This is an advantage when radioactive samples are of interest and the activity is a limiting factor (for example ^{48}V with activity of 0.17 mCi/ng). It is thus essential that detector development that will lead to experiments will also be done at the LANL LSDS.

2. Proposed Method

To develop new detection concepts, measurements of the reaction cross section will be done using a LSDS. Although measurements of small quantities of radioactive sample are of interest[ii], the high neutron flux will allow measurements in the range of millibarns on samples with quantities of only milligrams.

The samples of interest are ^{50}V, ^{64}Zn and ^{44}Ti and all have a positive *Q*-value which means that these are not threshold reactions and can thus be measured in the LSDS which

provide usable neutron flux in the energy range from 0.1 eV to 100 keV. Table I provides some information on the samples of interest.
The recent fission cross section measurements[i] at the LANL LSDS using a ^{239}Pu sample demonstrated that at 1 keV, a sample size of 30 ng was sufficient to measure a 10 barn cross section with accuracy of about 10% in 3 hours. Using the thermal cross section as an estimate about 90 mg should be sufficient for the (n,p) cross section of ^{50}V which is expected to be about 0.7 millibarns.

Table I - Properties of the samples of interest for (n,p) measurements. The proton kinetic energy is calculated assuming the neutron kinetic energy is zero.

Sample (Reaction)	Q [MeV]	Alpha/Proton Energy [MeV]	Theoretical estimate of cross section [barn]	Estimated required sample mass [mg]
V-50 (n,p)	4.79	4.70	0.7	90
Zn-64 (an)	3.866	3.64	0.24	270
Ti-44 (n,p)	1.049	1.026	16	3
Ti-44 (an)	3.325	3.048	0.56	100

2.1. The Lead Slowing Down Spectrometer

The lead slowing down spectrometer is capable of delivering high neutron fluxes that allow accurate measurements of fission cross sections with a fraction of a microgram sample of fissionable material. Currently there are only two lead-slowing-down-spectrometers in the USA: one is located at RPI and a recent smaller device is now located at LANL. The RPI LSDS will be used during the detector development and testing phase included in this proposal. The LANL LSDS will be later used to perform the actual measurement because of its proximity to the isotope production facility and its higher neutron flux. The LANL LSDS has about 2 order of magnitude higher neutron flux intensity compared to the RPI LSDS and thus allow measurements of smaller samples (or smaller cross sections)[i].
The RPI lead slowing down spectrometer, also known as the RPI Intense Neutron Spectrometer (RINS), is a 75-ton cube of pure lead, 1.8 meter on a side that is coupled to the RPI 100 MeV electron linear accelerator; this is the first high intensity LSDS spectrometer installed in the United States, where extensive experimental experience was gained. A schematic view of the RPI LSDS is shown in Figure 1. The electron beam hits an air-cooled tantalum target in the center of the assembly. The detector will be located inside the lead cube 60 cm from the lead cube face (see Figure 1).
The lead slowing down spectrometer was constructed as a tool for non-destructive assay of spent fuel[iii]. The availability of a high neutron flux spectrometer spawned extensive work on measurements of fission cross sections of radioactive samples using this facility[iv,v,vi]. The high flux is a result of the neutron slowing down process in lead. In this process the neutron loses energy by elastic collision with the lead. As a result of these collisions the neutrons tend to focus to an approximate Gaussian energy distribution where the average of the distribution E (eV) is decreasing as a function of slowing down time t (μs) according to

$$E = \frac{165000}{(t + 0.3)^2} . \quad (1)$$

The neutron flux as a function of neutron energy for a lead slowing down spectrometer driven by the LINAC delivering 1 kw of electron beam power to a neutron producing tantalum target is shown in Figure 2. This neutron flux is about 10^3-10^4 times higher than the neutron flux at a 5 meter time-of-flight experiment.

The high flux allows accurate measurement of about 1 barn cross section of a microgram sample. This sensitivity is a great advantage when measuring properties of fission fragments since the sample must be thin enough to allow the fission fragments to escape. Typical sample thickness is of the order of few tens of micrograms per cm^2. With the RINS a total sample size of the order of 100-200 nanograms can easily be measured.

In previous experiments we compared results obtained with the RINS and WNR at LANL and obtained excellent agreements for the measurements of the fission cross sections of radioactive samples[vii].

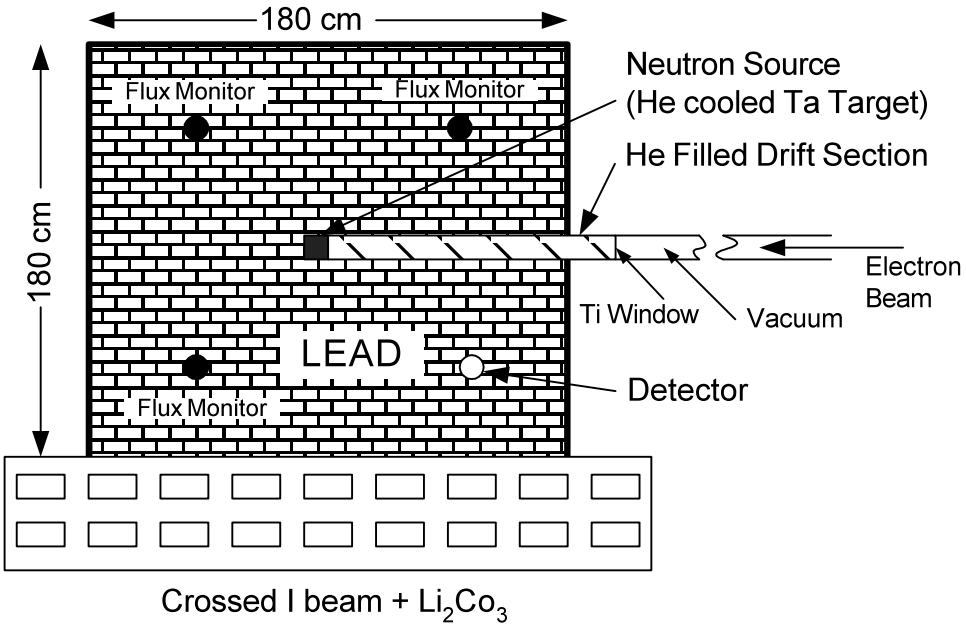


Figure 1 – A schematic view of the RPI lead slowing down spectrometer.

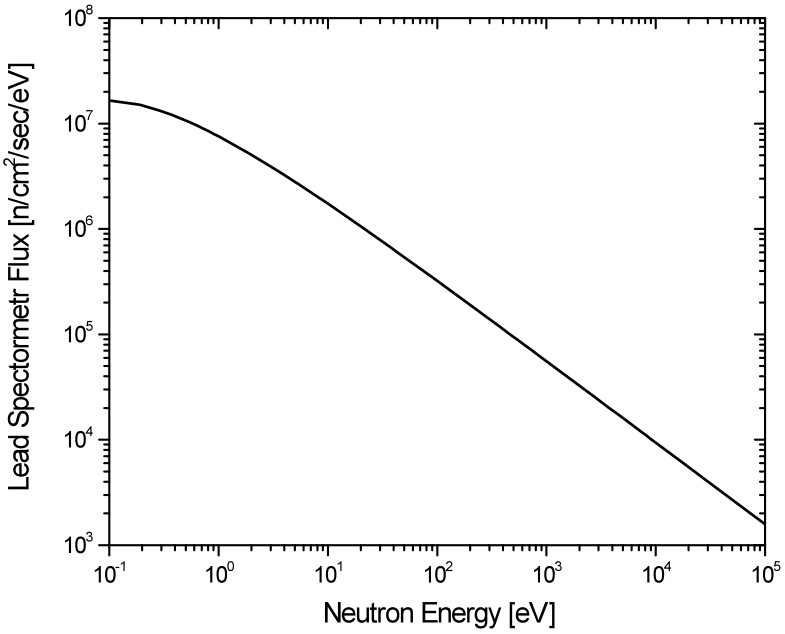


Figure 2 – The neutron flux as a function of energy inside a lead-slowing-down-spectrometer.

The approximate Gaussian energy distribution obtained by the neutrons when they slow down in energy determines the energy resolution of the RINS. In order to maximize the resolution, care was taken in assembling the spectrometer from 99.99% pure lead with minimal void and minimal hydrogen content in the lead assembly. The obtained resolution is about 35% FWHM in the broad neutron energy range of 0.1 eV – 50 keV. In some cases such as ^{239}Pu where the level spacing is large, information can be obtained on isolated resonances. Examples of the RINS capabilities to measure fission cross-sections of sub microgram quantities of short-lived isotopes are show in Figure 3. These measurements were the first to be done on these isotopes in this energy range. Figure 3 shows the fission cross-section of ^{247}Cm, ^{254}Es and ^{250}Cf. The ^{247}Cm measurement shows the ability of the RINS to resolve low energy resonances and higher energy resonance clusters. The ^{250}Cf sample had a large background due to spontaneous fission (442 fissions per second) and demonstrates the high signal to background ratio that enabled us to be the first to observe a resonance at 0.53 eV. The ^{254}Es sample had an activity of 0.4 mCi mostly in the form of alpha emission.

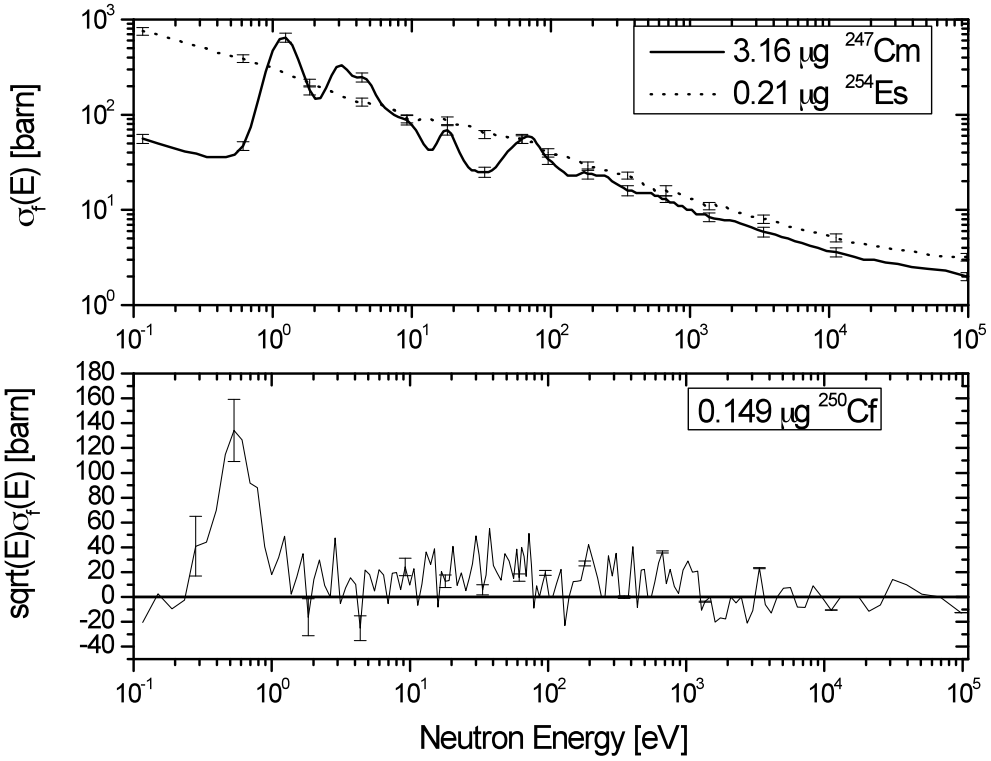


Figure 3 – Fission cross section of radioactive samples of ^{247}Cm, ^{254}Es and ^{250}Cf measured with the RINS[iv]. The activity of the ^{254}Es was about 0.4 mCi.

2.2. The Detector

There are several requirements for a detector for such measurements.

1. Provide a distinct signal from the alphas emitted during the reaction.
2. Have a fast recovery from the strong gamma flash inside the LSDS.
3. Has to fit inside the limited space of the LSDS and should contain as little hydrogenous materials as possible (which can degrade the neutron energy resolution).
4. Have a fast response to provide accurate slowing-down-time information.
5. Be capable of working in the background radiation inside the LSDS.

It is clear that requirement 1 is essential in order to get any information on the (n,α) reaction in the sample. The most demanding requirement is 2. Requirement 2 is a common problem for detectors used inside a LSDS and can be solved by careful design of an intrinsically-compensated detector which will minimize the effect of the gamma flash and provide fast detector recovery. For fission measurements this is typically achieved by using a compensated fission chamber or a compensated solid state detector such as a solar cell which was recently developed by the LSDS collaboration at LANL[viii].

2.2.1. Detector Designs

There are two detector designs that will be used. The design can be easily modified to expand the capability of these detectors to other types of experiment, for example (n,p) measurements. Also fine tuning of the design (mostly the dimensions) to the expected energy of the charge particles and the background radiation may need to be necessary. We have considered many possible designs and arrived at two working detectors based on: (1) a solid state Canberra Passivated Implanted Planar Silicon (PIPS) detector and (2) Gas-Electron-Multiplication (GEM) Foil.

Solid state detector

A solid state detector such as a solar cell was successfully used for fission measurements in the LANL LSDS. Fission fragments are relatively heavy and are emitted with high kinetic energy of tens of MeV, which is high compared to alphas of several MeV that are emitted from the neutron absorption reaction. The range of the alpha is much longer than the range of the fission fragments and thus the detector has to have a larger active thickness. A solar cell that works well for fission fragments might not work for these lighter and lower energy particles which are providing signals at the noise level of this detector.

Another possibility is a surface barrier detector. This type of detector is available with a variety of depletion layer thicknesses which can be selected to match the range of the alpha particles. For protons at 5 MeV a depletion layer of about 200 μm is required in order to fully stop the protons. Surface barrier detectors are sensitive to low energy gamma rays and some of our previous experience with these detectors inside the RPI LSDS was not as good as the detector exhibited gain drifts. However these experiments were done 30 years ago and since then semiconductor detectors continued to be improved. More recent experiments performed last year with a Passivated Implanted

Planar Silicon (PIPS) have proven that these new semiconductor detectors have improved enough to make then viable options. Figure 4 shows experimental results that indicate no permanent gain shifts or lose of resolution after exposure in the RPI LSDS. Figure 5 is a cross section measured for the (n,α) reaction on ^{6}Li, taken with a PIPS detector proving that this reaction is detectable in the LSDS environment. The detector was not compensated and only recovered from the gamma flash after the average neutron energy had already been reduced to 1 keV. Compensation should improve the recovery time and thus raise measured neutron the energy.

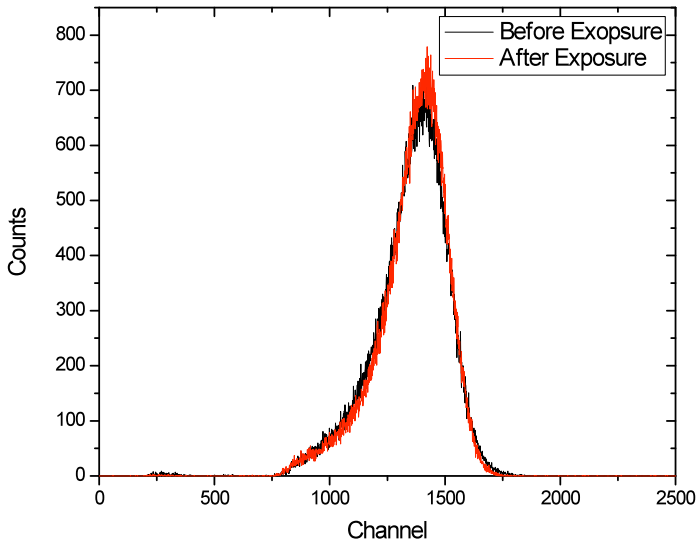


Figure 4 – Multi Channel Analyzer (MCA) spectrum of the PIPS detector using a Gd-148 α source (E_{α}=3.18MeV) shows that after about 4 hours in the RPI LSDS operating at 8 μA no detectible damage (gain shifts or lose of energy resolution) was done.

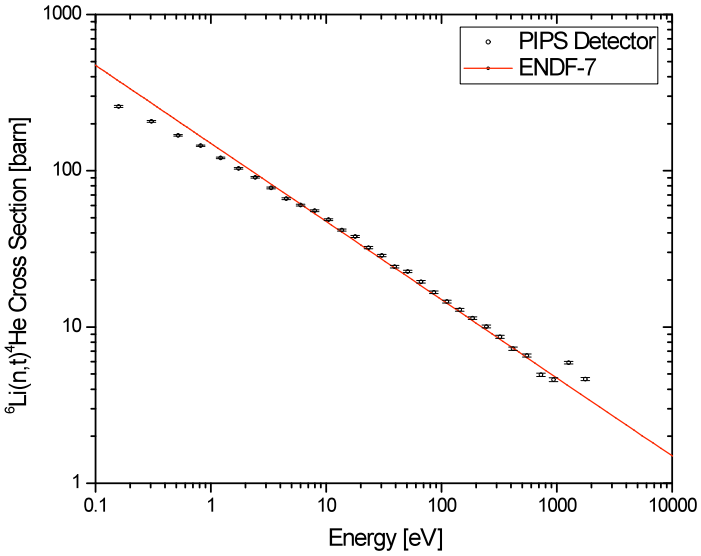


Figure 5 – (n,α) cross section measurements of ^{6}Li performed in the RPI LSDS using an uncompensated PIPS Detector

GEM Foil detector

The Gas Electron Multiplier (GEM) detector operates on the same principle as a proportional counter. A GEM foil is made of 50 µm thick polyamide or Kapton between two 5 µm thick copper layers. The entire foil is perforated with 30 µm holes. The GEM detector is constructed with the sample placed on the cathode allowing the alpha to traverse a few millimeters depositing energy in the form of ionizing the fill gas. The electrons are pulled towards the GEM foil. By applying a250-500 V potential, the electrons are forced though the high electric field in the vicinity of the holes where they are accelerated fast enough to cause secondary ionizations as they pass through the holes. After leaving the foil the electrons are drawn to the anode where the signal is extracted. Additional GEM foils can be placed in series for greater signal amplification.

GEM detectors have several advantages over the more traditional proportional counters which will be crucial in successfully performing (n,α) measurements. The small spacing allows for quick charge collection creating much faster signals, lower sensitivity to and quicker recovery from the gamma flash. Each GEM foil acts to shield the anode from the signal generated by the positive ions much like a Frisch grid.

Such detectors have already be designed for the LANL LSDS and tested at RPI. The third generation RPI-GEM detector is currently being constructed and will be ready for testing by early summer. The GEM detector also tested earlier this year is currently at about the same level as the PIPS detector as far as signal to noise ratio and recovery time. The Gen. II GEM detector is an uncompensated single foil modal which was able to handle count rates much larger than expected during the measurement of ^{50}V during a ^{6}Li(n,α) cross section measurement, see Figure 6.

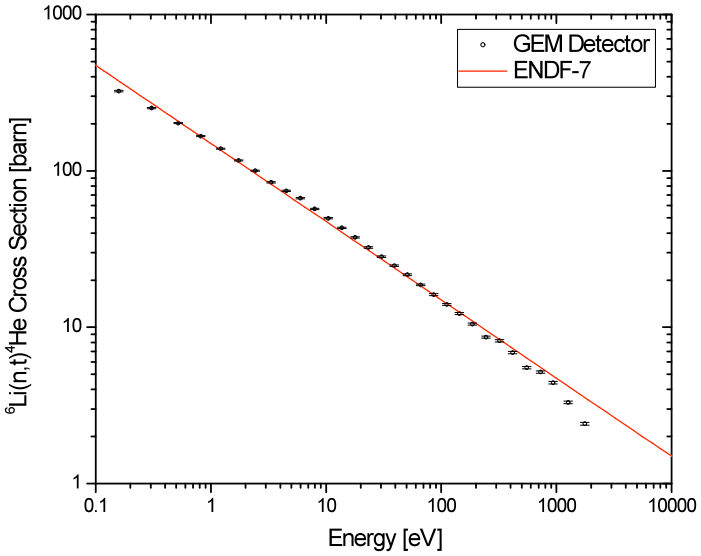


Figure 6 – (n,α) cross section measurements of ^{6}Li performed in the RPI LSDS using an uncompensated GEM Detector

3. Requirements

This measurement will require 1 week of use for the Blue Room Target 2, the LSDS. Day 1 will be used as setup and troubleshooting with or without the beam. Day 2 will entail testing of the equipment with the beam on, measuring detector responses to the γ-flash and other sources of background and using LiF to get a measurement of the flux and as a final check of the detectors operating status. Days 3-5 will be used to perform the actual measurements provided that we can obtain suitable sample.

4. Personnel

This research is directed by Dr. Danon who is an associated professor and director of the Gaerttner LINAC laboratory at Rensselaer Polytechnic Institute. Graduate students working on this project are Jason Thomson who is developing these capabilities as part of his PhD thesis. Other graduate and undergraduate students from RPI will help.

5. References

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